

Quantum corrections to the spin-wave spectrum of La_2CuO_4 in an external magnetic field

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Abstract

The effects of quantum fluctuations on the magnetic field dependence of the spin-wave gaps in the lamellar antiferromagnet La_2CuO_4 are considered. Nonlinear corrections to the spin-wave spectrum are calculated to leading order in $1/S$, where S is the localized spin. The nearest-neighbor exchange interactions between the Cu spins as well as the Dzyaloshinskii-Moriya interactions are taken into account. Using the experimental values of the components of the g -factor tensor, we get a satisfactory agreement with the experimental results for the field dependence of the gaps by Gozar *et al.* [Phys. Rev. Lett. **93**, 027001 (2004)], and obtain consistent values of the in-plane and inter-plane coupling constants. The field dependence of the dispersion of spin waves propagating perpendicular to the CuO_2 planes is also discussed.

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I. INTRODUCTION

Lanthanum cuprate, La_2CuO_4 , the parent compound of the high-temperature superconductors, is a layered weakly orthorhombic antiferromagnet (AF), with a Néel temperature, T_N , of approximately 325 K.¹ The orthorhombic distortion is associated with a small tilt of oxygen octahedra around each copper ion, which also introduces the antisymmetric Dzyaloshinskii-Moriya (DM) superexchange interaction between neighboring spins. Because of the DM interaction, each CuO_2 plane acquires a small ferromagnetic moment along the c -axis, perpendicular to the plane. The direction of the ferromagnetic moments alternates in adjacent planes, and, therefore, there is no net ferromagnetic moment in the crystal.

The DM interaction, though small, has a strong impact on the magnetic properties of La_2CuO_4 . It was established that a magnetic field perpendicular to the CuO_2 plane causes a first order weak-ferromagnetic transition (WFT).^{2,3,4} The critical field, H_c , of the transition depends on the DM coupling as well as on the interlayer exchange. In a non-stoichiometric sample ($T_N = 234$ K), studied in Ref. 3, the critical field at low temperatures was $H_c \approx 4.8$ T. In Ref. 5 the value $H_c \approx 6$ T was obtained from Raman spectrum measurements for a crystal with $T_N = 310$ K, while in a recent paper⁶ a significantly larger transition field of 11.5 T was found from neutron diffraction studies for a sample with $T_N = 316$ K.

Another manifestation of the DM interaction in La_2CuO_4 is the phase diagram in an in-plane magnetic field. The antiferromagnetic staggered moment in La_2CuO_4 is directed along the diagonal of the CuO plaquette (b -axis), the in-plane anisotropy being generated by the DM interaction. A theory⁷ based on the mean-field version of the Hamiltonian, which includes exchange and DM coupling between Cu spins, as well as an out-of-plane anisotropy term, predicts that in a field along the b -axis one should observe two phase transitions: a spin-flop transition, at which the staggered moment jumps from the (b,c) plane into the (b,a) plane (**a** is perpendicular to **b** and **c**) at a field H , determined by the DM coupling energy, and a second transition at a higher field, when the staggered moment rotates into the c -direction. Such a phase diagram was indeed observed in Ref. 7. Note, however, that in Refs. 5,6,8 no spin-flop transition was observed. According to these references, the staggered moments rotate gradually from the b - to the c -axis.

Gozar *et al.*⁵ used Raman spectroscopy measurements to study the influence of a magnetic field on the in-plane spin-wave (SW) gap in La_2CuO_4 . They showed that the gap abruptly

increases at the WFT, decreases with the increase of the field directed along the b -axis and increases with the field pointing along the a -axis. The observed field dependence of the gaps was described by classical theories^{9,10,11,12} based on the model, which include anisotropic symmetric and antisymmetric intralayer coupling between the Cu spins as well as interlayer exchange.

On the other hand, it is known that quantum corrections are very important when considering the properties of layered cuprates, with spin $S = 1/2$.¹³ Intensive calculations were performed for an isotropic Heisenberg two dimensional model with nearest neighbor interactions.^{13,14,15,16,17,18,19} Perturbative expansions in $1/S$, series expansions, and Monte Carlo methods were used to go beyond the linear SW theory.^{13,16,18} It appears that the renormalization factors obtained from the nonlinear SW (NLSW) theory to leading order in $1/S$ are sufficiently close to those which follow from more complicated calculations even for spin $S = 1/2$. The quantum effects renormalize significantly the mean value of the spin at $T = 0$, the SW stiffness, the in-plane and out-of-plane SW gaps at zero magnetic field, etc. In a recent paper,²⁰ the $1/S$ NLSW theory was employed to calculate the effect of quantum fluctuations on the SW stiffness and other parameters of La_2CuO_4 , in a model which takes into account the plaquette ring exchange. The renormalization factors which follow from these calculations and the fit of the experimental results by Coldea *et al.*,²¹ appeared to be somewhat closer to unity than in the model with nearest-neighbor exchange only. In view of these results one should expect that quantum fluctuations are important also for the quantitative understanding of the SW spectrum behavior in a magnetic field.

In this paper we use the NLSW theory to leading order in $1/S$ to calculate the influence of quantum fluctuations on the magnetic field dependence of the in-plane and out-of-plane SW gaps in La_2CuO_4 . We show that the quantum fluctuations alter the expression for the critical field of the WFT, renormalize all physical quantities, which enter in the equations for the gaps in a magnetic field, and change therefore significantly the values of the spin-spin couplings extracted from the experiment. The theoretical results are in satisfactory agreement with the experimental findings. The theory also predicts a non-trivial effect of the magnetic field on the SW propagating in the c -direction.

The paper is organized as follows. In section II we present the model of magnetic interactions in La_2CuO_4 used in our calculations. In section III we derive the field dependence of the gaps for the field in the c -direction in both the linear and the nonlinear SW approxi-

mations. We also consider the field dependence of the dispersion of SW, which propagate in the c -direction. In section IV the NLSW theory for the SW gaps in a magnetic field along the b - and a -directions is developed. In section V we compare our NLSW results with the experiments by Gozar *et al.*⁵ Finally in the Appendix the Green's function are used to derive a general expression for the eigenvalue matrix of a model described by the Hamiltonian of the type (14). The Appendix also presents some useful relations between the averages of the magnon operators.

II. THE MODEL

Our Hamiltonian is written as²²

$$\begin{aligned} \mathcal{H} = & J \sum_{\langle ij \rangle} [\mathbf{S}_i \cdot \mathbf{S}_j - \alpha S_i^z S_j^z] + \sum_{\langle ij \rangle} \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j \\ & + J_{\perp} \sum_{\langle ik \rangle} \mathbf{S}_i \cdot \mathbf{S}_k - \sum_i \mu_B \mathbf{H} \cdot \hat{\mathbf{g}} \cdot \mathbf{S}_i. \end{aligned} \quad (1)$$

Here the first term describes the anisotropic exchange coupling between nearest-neighbor spins in the CuO_2 layers, the easy plane being the crystallographic (a, b) plane. The anisotropy factor α in La_2CuO_4 (LCO) is small, of order 10^{-4} .²³ The second term is the DM interaction, which leads to the canting of the spins out of the plane. The third term is the small interlayer exchange coupling. The sum is over the (two) nearest neighbors along the crystallographic c -axis, which we choose as the z -axis. Finally, the last term in Eq. (1) describes the Zeeman energy in an external field \mathbf{H} , $\hat{\mathbf{g}}$ being the anisotropic g -factor; μ_B is the Bohr magneton.

The DM vector \mathbf{D}_{ij} is perpendicular to the Cu-Cu bonds and changes sign from one bond to the next one. In the orthorhombic crystallographic axes (a, b, c) , where \mathbf{a} and \mathbf{b} point along the diagonals of the Cu plaquette, the vector \mathbf{D}_{ij} can be written as

$$\begin{aligned} \mathbf{D}_{ii+\delta_1} &= (D, D, 0) \\ \mathbf{D}_{ii+\delta_2} &= (D, -D, 0), \end{aligned} \quad (2)$$

where δ_1 and δ_2 are directed along the two orthogonal bonds of the plaquette. Using these DM vectors, the DM term in the Hamiltonian transforms into

$$\sum_{\langle ij \rangle} \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j = \sum_{\langle ij \rangle} \mathbf{D} \cdot \mathbf{S}_i \times \mathbf{S}_j$$

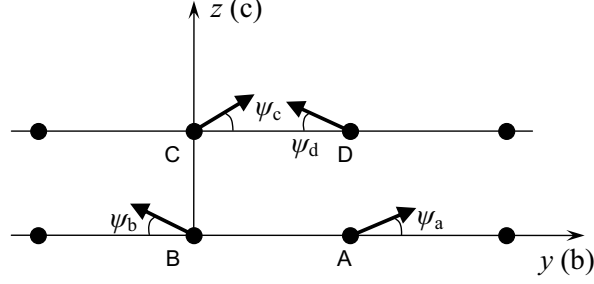


FIG. 1: The four copper spins A,B,C,D in a unit cell. Ordered spin directions are shown by arrows. The horizontal lines refer to the CuO_2 layers. The global reference axes z and y point along the orthorhombic crystallographic axes c and b respectively.

$$+ D \sum_i \left[\sum_{\delta_1} \left(S_i^x S_{i+\delta_1}^z - S_i^z S_{i+\delta_1}^x \right) - \sum_{\delta_2} \left(S_i^x S_{i+\delta_2}^z - S_i^z S_{i+\delta_2}^x \right) \right], \quad (3)$$

with the vector \mathbf{D} of length D pointing along \mathbf{a} . The axes (x, y, z) are along (a, b, c) .

The second term in the r.h.s. of this expression does not contribute to the gap. It only changes slightly, to order D/J , the SW dispersion.²⁴ We will therefore neglect it in what follows. Thus, for our purposes the DM interaction can be described by the vector \mathbf{D} , directed along the a -axis in each plaquette.

The magnetic unit cell in the above model contains four spins, A, B, C, D , in two inequivalent layers, Fig. 1. Due to the DM interaction, these spins cant into the c -direction, with canting angles, ψ_μ , with $\mu = a, b, c, d$ for the spins in the unit cell. These angles depend on the direction and on the strength of the magnetic field. In zero field, they satisfy the relation $\psi_a = \psi_b = -\psi_c = -\psi_d$, so that the magnetic moment of the unit cell is equal to zero. To calculate the SW spectrum, it is convenient to rotate the spins from the above global crystallographic axes (x, y, z) to local axes (ξ, η, ζ) , with ζ along the sublattice magnetization. When the magnetic field is in the b -direction, and smaller than the spin-flop field, or in the c -direction, the spins rotate in the (y, z) plane. Then the transformation equations from the crystallographic axes to the local ones for spins a and b in the sublattices A and B are (see Fig. 1)

$$\begin{aligned} S_a^y &= S_a^\eta \sin \psi_a + S_a^\zeta \cos \psi_a, & S_a^z &= -S_a^\eta \cos \psi_a + S_a^\zeta \sin \psi_a, \\ S_b^y &= S_b^\eta \sin \psi_b - S_b^\zeta \cos \psi_b, & S_b^z &= S_b^\eta \cos \psi_b + S_b^\zeta \sin \psi_b. \end{aligned} \quad (4)$$

Analogous equations hold for the spins c and d in the second layer.

III. FIELD ALONG THE c -AXIS

The canting angles for this direction of the field are the same for each pair of the spins in a plane, i.e. $\psi_a = \psi_b \equiv \psi_1$, and $\psi_c = \psi_d \equiv \psi_2$. ψ_1 and ψ_2 , and, hence, the magnetic moments of the planes differ if H is smaller than a critical field H_c , which in the classical approximation is equal to²

$$H_c = \frac{4JJ_\perp S}{g_c\mu_B D}. \quad (5)$$

At $H = H_c$ the weak-ferromagnetic transition (WFT) happens, and the magnetic moments of all planes became the same.² At fields of order H_c , the angles ψ_1 and ψ_2 are small, of order 10^{-2} .

In the local framework, the Hamiltonian (1) has the form

$$\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_\perp. \quad (6)$$

Here

$$\begin{aligned} \mathcal{H}_{l=1,2} = & -(J \cos 2\psi_l + D_l \sin 2\psi_l + \alpha J \sin^2 \psi_l) \sum_{\langle ij \rangle} S^\zeta(i) S^\zeta(j) \\ & + \frac{1}{2} \left(J \cos^2 \psi_l + \frac{D_l}{2} \sin 2\psi_l - \frac{\alpha J}{2} \right) \sum_{\langle ij \rangle} [S^+(i) S^+(j) + S^-(i) S^-(j)] \\ & + \frac{1}{2} \left(J \sin^2 \psi_l - \frac{D_l}{2} \sin 2\psi_l + \frac{\alpha J}{2} \right) \sum_{\langle ij \rangle} [S^+(i) S^-(j) + S^-(i) S^+(j)] \\ & - g_c \mu_B H \sin \psi_l \left[\sum_i S^\zeta(i) + \sum_j S^\zeta(j) \right] + g_c \mu_B H \cos \psi_l \left[\sum_i S^\eta(i) - \sum_j S^\eta(j) \right] \\ & + \left(J \sin 2\psi_l - D_l \cos 2\psi_l - \frac{\alpha J}{2} \sin 2\psi_l \right) \sum_{\langle ij \rangle} [S^\zeta(i) S^\eta(j) - S^\eta(i) S^\zeta(j)], \end{aligned} \quad (7)$$

where $D_l = (-1)^l D$,

$$S^\pm = S^\zeta \pm i S^\eta, \quad (8)$$

and $i(j)$ labels the spins in the sublattice A (B).

The interlayer Hamiltonian transforms into

$$\begin{aligned} \mathcal{H}_\perp = & -J_\perp \sum_{\langle ik \rangle} \left\{ S^\zeta(i) S^\zeta(k) \cos \psi_{1,2} - \frac{1}{2} [S^+(i) S^+(k) + S^-(i) S^-(k)] \cos^2 \frac{\psi_{1,2}}{2} \right. \\ & \left. - \frac{1}{2} [S^+(i) S^-(k) + S^-(i) S^+(k)] \sin^2 \frac{\psi_{1,2}}{2} + [S^\zeta(i) S^\eta(k) - S^\eta(i) S^\zeta(k)] \sin \psi_{1,2} \right\}, \end{aligned} \quad (9)$$

where $\psi_{1,2} = \psi_1 + \psi_2$. We next make the Dyson-Maleev transformation to the bosons operators a, b, c, d for the four spin operators in a unit cell. In the local framework, this

transformation is the same for all spins. It can be written as

$$\begin{aligned} S_\mu^+ &= \sqrt{2S}\alpha_\mu, \quad S_\mu^- = \sqrt{2S}\alpha_\mu^\dagger \left(1 - \frac{\alpha_\mu^\dagger \alpha_\mu}{2S}\right), \\ S_\mu^z &= S - \alpha_\mu^\dagger \alpha_\mu, \end{aligned} \quad (10)$$

where the operators α_μ are defined as $\alpha_1 = a$, $\alpha_2 = b$, $\alpha_3 = c$, $\alpha_4 = d$.

A. Linear spin-wave theory

It follows from Eqs (6), (7), (9) that the classical ground-state energy per spin, E_0 , is

$$\begin{aligned} E_0 &= \frac{1}{4}[-JS^2z(\cos 2\psi_1 + \cos 2\psi_2) + DS^2z(\sin 2\psi_1 - \sin 2\psi_2) \\ &\quad - 2g_c\mu_B HS(\sin \psi_1 + \sin \psi_2) - 4J_\perp S^2 \cos(\psi_1 + \psi_2) - \alpha JS^2z(\sin^2 \psi_1 + \sin^2 \psi_2)], \end{aligned} \quad (11)$$

where $z = 4$ is the number of nearest neighbors in the plane. The minimization conditions for the energy E_0 with respect to ψ_1 and ψ_2 yield

$$\begin{aligned} JSz(2 - \alpha) \sin 2\psi_1 + 4J_\perp S \sin(\psi_1 + \psi_2) + 2DSz \cos 2\psi_1 - 2g_c\mu_B H \cos \psi_1 &= 0, \\ JSz(2 - \alpha) \sin 2\psi_2 + 4J_\perp S \sin(\psi_1 + \psi_2) - 2DSz \cos 2\psi_2 - 2g_c\mu_B H \cos \psi_2 &= 0. \end{aligned} \quad (12)$$

The solution of these equations gives ψ_1 and ψ_2 at fields $H < H_c$. Given that ψ_1 and ψ_2 are small, and neglecting terms of order α and J_z/J , we get

$$\psi_1 = \frac{g_c\mu_B H - DSz}{2JSz}, \quad \psi_2 = \frac{g_c\mu_B H + DSz}{2JSz}. \quad (13)$$

With these angles, the terms in Eq. (7) which are proportional to S^η , and are therefore linear in the boson operators, cancel out.

The bilinear spin-wave Hamiltonian, which follows after the transformation (10) is performed, can be written as

$$\mathcal{H}^{(2)} = \sum_{\mathbf{q}} \left\{ A_{\mu\nu}(\mathbf{q}) \alpha_\mu^\dagger(\mathbf{q}) \alpha_\nu(\mathbf{q}) + \frac{1}{2} B_{\mu\nu}(\mathbf{q}) [\alpha_\mu^\dagger(\mathbf{q}) \alpha_\nu^\dagger(-\mathbf{q}) + \alpha_\mu(\mathbf{q}) \alpha_\nu(-\mathbf{q})] \right\}, \quad (14)$$

where the matrices $\mathbf{A}(\mathbf{q})$ and $\mathbf{B}(\mathbf{q})$ are given by

$$\begin{aligned} A_{11} &= A_{22} = JSz(1 - 2R_1) + g_c\mu_B H \psi_1 + 2J_\perp S, \\ A_{33} &= A_{44} = JSz(1 - 2R_2) + g_c\mu_B H \psi_2 + 2J_\perp S, \end{aligned}$$

$$\begin{aligned}
A_{12} &= A_{21} = JSz\gamma_{\mathbf{q}}\left(R_1 + \frac{\alpha}{2}\right), \\
A_{34} &= A_{43} = JSz\gamma_{\mathbf{q}}\left(R_2 + \frac{\alpha}{2}\right), \\
A_{14} &= A_{23} = A_{13} = A_{14} = 0, \\
B_{12} &= B_{21} = JSz\gamma_{\mathbf{q}}\left(1 - R_1 - \frac{\alpha}{2}\right), \\
B_{34} &= B_{43} = JSz\gamma_{\mathbf{q}}\left(1 - R_2 - \frac{\alpha}{2}\right), \\
B_{14} &= B_{23} = 2J_{\perp}Sc_z, \\
B_{11} &= B_{22} = B_{33} = B_{44} = B_{13} = B_{31} = B_{24} = B_{42} = 0,
\end{aligned} \tag{15}$$

while

$$R_{l=1,2} = \psi_l\left(\psi_l - \frac{D_l}{J}\right) = \frac{H^2 - (DSz)^2}{4(JSz)^2}, \tag{16}$$

and

$$\gamma_{\mathbf{q}} = \frac{\cos q_x + \cos q_y}{2}, \quad c_z = \cos q_z. \tag{17}$$

The wave-vector components in the plane and along the z -axis are measured in units of the corresponding intersite distances. In Eq. (15) we neglect corrections of order $J_{\perp}\psi_l^2$ and $\alpha\psi_l^2$ ($l=1,2$).

It was shown in Ref. 19 (see also the Appendix of this paper) that the squares of the spin-wave energy, ω^2 , are the eigenvalues of the matrix

$$\mathbf{M} = [\mathbf{A}(\mathbf{q}) + \mathbf{B}(\mathbf{q})] \times [\mathbf{A}(\mathbf{q}) - \mathbf{B}(\mathbf{q})]. \tag{18}$$

Since the magnetic field is small, $g_c\mu_B H \ll J$, it is not expected to influence essentially the stiffness of the spin-waves propagating in the (x, y) plane. In contrast, the effect of the field on the spin waves propagating in the z -direction may be strong. We consider, therefore, only the dispersion of the spin waves propagating in this direction, and put $\gamma_{\mathbf{q}} = 1$. The spin-wave spectrum at $H < H_c$, which follows then from Eqs. (15) and (18), is given by

$$\begin{aligned}
\omega_{in}^2 &= \delta^2 + w^2 \pm \sqrt{(g_c\mu_B H \delta)^2 + w^4 c_z^2}, \\
\omega_{out}^2 &= \Delta^2 + (g_c\mu_B H)^2 + w^2 \pm \sqrt{(g_c\mu_B H \delta)^2 + w^4 c_z^2}.
\end{aligned} \tag{19}$$

Here ω_{in} and ω_{out} are the frequencies of the in-plane and out-of-plane spin waves, respectively. The zero-field in-plane, δ , and out-of-plane, Δ , gaps are:

$$\delta = DSz, \quad \Delta = JSz\sqrt{2\alpha}, \tag{20}$$

and $w^2 = 4zJJ_\perp S^2$. The lower (upper) sign in Eqs. (19) gives the frequencies of the in-phase or acoustical (out-of-phase or optical) oscillations in adjacent planes.

At $H = H_c$, the field which favors alignment of magnetic moments in adjacent planes overcomes the interlayer coupling which favors alternation of the moments. Then the staggered moments of all the planes align in the same direction, accompanied by a 180° rotation of the spins in half of the planes. Hence, the canting angles at $H > H_c$ are

$$\psi_2 = \pi - \psi_1 = \frac{g_c \mu_B H + DSz}{2JSz}. \quad (21)$$

The difference ΔE_0 , between the ground state energy E_0 at $H < H_c$ and that at $H > H_c$, is

$$\Delta E_0 = \frac{g_c \mu_B H DS}{2J} - 2J_z S^2. \quad (22)$$

The expression (5) for H_c follows from the relation $\Delta E_0 = 0$.

Since the two adjacent planes are equivalent, only the two modes which describe the in-phase oscillations in the planes are relevant. One gets for these modes

$$\begin{aligned} \omega_{in}^2 &= \delta^2 + g_c \mu_B H \delta + w^2(1 - c_z), \\ \omega_{out}^2 &= \Delta^2 + (g_c \mu_B H)^2 + g_c \mu_B H \delta + w^2(1 - c_z). \end{aligned} \quad (23)$$

The spin-wave gaps which follow from Eqs. (19) and (23) at $c_z = 1$, coincide with those obtained in different ways in Refs. 9-12.

B. Nonlinear spin waves

We now consider the effect of terms which are cubic and quartic in the boson operators. It follows from Eqs. (7) and (9) that all the terms which are linear in S^η , except those which are proportional to H , cancel out owing to Eq. (12). Thus, the only terms odd (cubic) in boson operators that remain in the Hamiltonian are proportional to H . For one plane and for small fields $g_c \mu_B H \ll JSz$ we have

$$\mathcal{H}^{(3)} = g_c \mu_B H \sqrt{\frac{1}{2S}} \frac{1}{z} \sum_{\langle ij \rangle} [a_i^\dagger a_i (b_j - b_j^\dagger) + b_j^\dagger b_j (a_i^\dagger - a_i)]. \quad (24)$$

The fourth-order terms of order $1/S$ come from the exchange and DM interactions, and from the out-of-plane anisotropy. They are

$$\mathcal{H}_J^{(4)} = -J(1 - 2\psi_1^2) \sum_{\langle ij \rangle} a_i^\dagger a_i b_j^\dagger b_j - \frac{J}{2}(1 - \psi_1^2) \sum_{\langle ij \rangle} a_i^\dagger b_j^\dagger (a_i^\dagger a_i + b_j^\dagger b_j)$$

$$- \frac{J}{2} \psi_1^2 \sum_{\langle ij \rangle} a_i^\dagger a_i^\dagger a_i b_j, \quad (25)$$

$$\mathcal{H}_D^{(4)} = \frac{D\psi_1}{2} \sum_{\langle ij \rangle} [4a_i^\dagger a_i b_j^\dagger b_j + a_i^\dagger a_i^\dagger a_i (b_j^\dagger - b_j) + (a_i^\dagger - a_i) b_j^\dagger b_j^\dagger b_j], \quad (26)$$

$$\mathcal{H}_\alpha^{(4)} = -\frac{J\alpha}{4} \sum_{\langle ij \rangle} [(a_i - a_i^\dagger) b_j^\dagger b_j^\dagger b_j + (b_j - b_j^\dagger) a_i^\dagger a_i^\dagger a_i]. \quad (27)$$

The $1/S$ corrections to the spin wave spectrum come from the first-order contribution of $\mathcal{H}^{(4)}$ and the second-order contribution of $\mathcal{H}^{(3)}$. The last contribution is of order H^2 , and it renormalizes the H^2 terms in the out-of plane spin gap, Eqs. (19), (23). It was calculated in Ref. 25.

To treat the quartic perturbation, we truncate all four-operator terms by contracting out pairs of operators in all possible ways and discarding the non-Hermitian terms.¹⁹ One finally arrives at an effective bilinear Hamiltonian, of the form shown in Eq. (14), with the matrix elements $A_{\mu\nu}$ and $B_{\mu\nu}$ depending on the following averages of the magnon operators:

$$\begin{aligned} \nu &= \langle a_i^\dagger a_i \rangle = \sum_{\mathbf{q}} a_{\mathbf{q}}^\dagger a_{\mathbf{q}}, \quad \xi = \sum_{\mathbf{q}} \langle a_{\mathbf{q}} b_{-\mathbf{q}} \rangle = \gamma_{\mathbf{q}}, \\ \eta &= \sum_{\mathbf{q}} \langle a_{\mathbf{q}}^\dagger b_{\mathbf{q}} \rangle = \gamma_{\mathbf{q}}, \quad \lambda = \sum_{\mathbf{q}} \langle a_{\mathbf{q}} a_{-\mathbf{q}} \rangle. \end{aligned} \quad (28)$$

The matrix elements $A_{\mu\nu}$ and $B_{\mu\nu}$, with $\mu = 1, 2; \nu = 1, 2$ can be written as

$$\begin{aligned} A_{11} &= A_{22} = JSzZ_c \left[1 - 2\tilde{R}_1 \left(1 + \frac{\xi}{2S} \right) + \frac{\alpha\xi}{2S} \right] + H\tilde{\psi}_1 + 2J_z S Z_m, \\ A_{12} &= A_{21} = JSz\gamma_{\mathbf{q}} \left[\tilde{R}_1 Z_m - \frac{1}{2S} (\lambda + 2\eta) + Z_m \frac{\alpha}{2} \right], \\ B_{11} &= B_{22} = -\frac{Jz}{2} \left(\xi \tilde{R}_1 + \eta - \frac{\alpha\xi}{2S} \right), \\ B_{12} &= B_{21} = JSz\gamma_{\mathbf{q}} Z_c \left[1 - \tilde{R}_1 \left(1 - \frac{\xi}{S} \right) - \frac{\alpha}{2} \left(1 + \frac{\xi}{S} \right) \right]. \end{aligned} \quad (29)$$

Here $\tilde{\psi}_1$ is the tilting angle renormalized by quantum fluctuations (see below),

$$\tilde{R}_1 = \tilde{\psi}_1 \left(\tilde{\psi}_1 + \frac{D}{J} \right), \quad (30)$$

$Z_c = 1 - \nu/S - \xi/S$ is the renormalization of the SW velocity,^{13,26} and $Z_m = 1 - \nu/S$ is the renormalization factor of the average spin.

The equations for the matrix elements $A_{\mu\nu}$ and $B_{\mu\nu}$, with $\mu = 3, 4; \nu = 3, 4$ follow from the above equations by substituting $\tilde{\psi}_2$ for $\tilde{\psi}_1$. The mixed elements B_{14} and B_{23} are

$$B_{14} = B_{23} = 2J_\perp S Z_m c_z. \quad (31)$$

Note that averages like $\langle a_i c_j \rangle$ appear in the course of the renormalization. They are small, since i and j belong to different planes. When deriving Eqs. (29) we neglected also terms of order of $\eta\psi_n$ or $\lambda\psi_n$, since (as shown in the Appendix) λ and η are of order of ψ_1^2 .

The renormalization factors ξ and ν are known^{13,16} to be equal to

$$\nu = \frac{1}{2} \sum_{\mathbf{q}} \left(\frac{1}{\sqrt{1 - \gamma_{\mathbf{q}}^2}} - 1 \right) = 0.197, \quad \xi = -\frac{1}{2} \sum_{\mathbf{q}} \frac{\gamma_{\mathbf{q}}^2}{\sqrt{1 - \gamma_{\mathbf{q}}^2}} = -0.276. \quad (32)$$

This gives $Z_c = 1 + 0.158/2S = 1.158$. More accurate values of Z_c , obtained from expanding to order $1/S^2$ or from Monte Carlo simulations, are also available.^{16,18} Nevertheless, for consistency we use in what follows this value of Z_c . The factors η and λ are given in the Appendix. It appears that the spin-wave frequencies depend only on the sum $\eta + \lambda$, which according to Eqs. (29) and (A.10) is equal to

$$\lambda + \eta = -\left(R_1 + \frac{\alpha}{2}\right)\xi. \quad (33)$$

To calculate the renormalized tilting angle we add to the classical ground-state energy (11) the $(1/S)$ correction, which follows from the bilinear Hamiltonian (14) after averaging the products of the boson operators. This yields for the quantum corrected ground-state energy, E :

$$\begin{aligned} E = & \frac{1}{4} \left\{ \left(1 - \frac{2\nu + \xi}{S} \right) [-JS^2 z (\cos 2\psi_1 + \cos 2\psi_2) + DS^2 z (\sin 2\psi_1 - \sin 2\psi_2)] \right. \\ & \left. - 2g_c\mu_B HS \left(1 - \frac{\nu}{S} \right) (\sin \psi_1 + \sin \psi_2) - 4J_{\perp} S^2 \left(1 - \frac{2\nu}{S} \right) \cos(\psi_1 + \psi_2) \right\}. \end{aligned} \quad (34)$$

We neglected here terms of order $\eta\psi_1$, as well as small corrections which come from the out-of-plane anisotropy.

Minimization of the energy (34) gives

$$\tilde{\psi}_{2,1} = \frac{g_c\mu_B H}{2JZ_c S z} \pm \frac{D}{2J}, \quad (35)$$

in agreement with previous calculations for an AF with $D = 0$.^{27,28} Note that the contribution to the canting angle caused by the DM interaction is the same as in the classical limit.

When calculating the in-plane spin-wave energy from Eqs. (18) and (29), one finds a contribution to the gap which is of order $1/S$ and proportional to H^2 . This contribution

should be cancelled out by the contribution of the cubic terms in the Hamiltonian, since the H^2 term in the in-plane gap is forbidden by symmetry. Discarding this term, we get

$$\begin{aligned}\omega_{in}^2 &= \tilde{\delta}^2 + \tilde{w}^2 \pm \sqrt{(g_c\mu_B H)^2 \left(1 - \frac{\xi}{S}\right)^2 \tilde{\delta}^2 + \tilde{w}^4 c_z^2}, \\ \omega_{out}^2 &= \tilde{\Delta}^2 + Z_c(g_c\mu_B H)^2 + \tilde{w}^2 \pm \sqrt{(g_c\mu_B H)^2 \left(1 - \frac{\xi}{S}\right)^2 \tilde{\delta}^2 + \tilde{w}^4 c_z^2}.\end{aligned}\quad (36)$$

Here $\tilde{\delta} = Z_m\delta = (1 - \nu/S)\delta$, $\tilde{\Delta} = Z_m\Delta$, $\tilde{w} = Z_w w$, where $Z_w = 1 - \nu/S - \xi/2S \approx 0.88$. As noticed above, the renormalization of the H^2 term in the out-of-plane wave was calculated in Ref. 25.

We see that the quantum fluctuations effectively increase the g -factor in the c -direction. This is in contrast to the result,¹⁰ obtained in the limit of large N , N being the number of the components of the spin.

Above the WFT, when the staggered moments in all planes align in the same direction, the angle ψ_2 is given by Eq. (35), while ψ_1 is given by $\psi_1 = \pi - \psi_2$. It follows from Eq. (34) that the difference, ΔE in the ground state energy before and after the transition is

$$\Delta E = \frac{g_c\mu_B H S D}{2J} \left(1 - \frac{\nu}{S}\right) - 2J_\perp S^2 \left(1 - \frac{2\nu}{S}\right). \quad (37)$$

The condition $\Delta E = 0$ then yields the transition field,

$$H_c = \frac{4JZ_m J_z S}{g_c\mu_B D} = \frac{\tilde{w}^2}{\tilde{\delta}} \left(1 + \frac{\xi}{S}\right). \quad (38)$$

The above expression for H_c follows from the following simple qualitative arguments. At the WFT the magnetic moment increases abruptly by the value $\langle S_z \rangle = D/J = Z_m S D/J$ [see Eq. (35)], and the gain in the magnetic energy per spin is $\langle S_z \rangle g_c\mu_B H D/J$. The loss in the exchange energy is $2J_z \langle S_z \rangle^2$. Equating these energies one arrives at Eq. (38).

The SW spectrum at $H > H_c$ is

$$\begin{aligned}\omega_{in}^2 &= \tilde{\delta}^2 + H\tilde{\delta} \left(1 - \frac{\xi}{S}\right) + \tilde{w}^2(1 - c_z), \\ \omega_{out}^2 &= \tilde{\Delta}^2 + Z_c H^2 + H\tilde{\delta} \left(1 - \frac{\xi}{S}\right) + \tilde{w}^2(1 - c_z).\end{aligned}\quad (39)$$

IV. FIELD IN THE (a, b) PLANE

A. Field along the b -axis

When a magnetic field is applied in the direction of the staggered magnetization, i.e. along the b -axis, an unusual spin-flop transition happens at the field $H = \delta$.⁷ At this field the staggered moments rotate from the (b, c) plane to the (a, c) one, forming an angle with the c -axis, which decreases with the increase of the field. At a higher field, equal to $H = (\Delta^2 + 2w^2 - \delta^2)/\delta$, the staggered magnetization points in the c -direction.⁷

We consider in this paper only fields smaller than δ , when the spins lie in the (b, c) plane, which we defined as the (y, z) plane. As in the case of a field along the c -axis, the DM interaction and the field cause the canting of the spins out of the (a, b) plane. The canting angles of the spins a and b , however, are no longer equal to each other.

After the rotation to the local framework is performed, we obtain the Hamiltonian for the first plane (Fig. 1) as follows:

$$\mathcal{H}_1 = \mathcal{H}_{ev} + \mathcal{H}_{odd}. \quad (40)$$

Here

$$\begin{aligned} \mathcal{H}_{ev} = & -J \cos \psi_{ab} \sum_{\langle ij \rangle} S^\zeta(i) S^\zeta(j) + \frac{J}{2} \cos^2 \frac{\psi_{ab}}{2} \sum_{\langle ij \rangle} [S^+(i) S^+(j) + S^-(i) S^-(j)] \\ & + \frac{J}{2} \sin^2 \frac{\psi_{ab}}{2} \sum_{\langle ij \rangle} [S^+(i) S^-(j) + S^-(i) S^+(j)] + g_b \mu_B H (\cos \psi_b - \cos \psi_a) \sum_i S^\zeta(i) \\ & + D \sin \psi_{ab} \sum_{\langle ij \rangle} [S^\zeta(i) S^\zeta(j) - \frac{1}{4} (S^+(i) - S^-(i)) (S^+(j) - S^-(j))] \\ & - \frac{\alpha J}{4} \sum_{\langle ij \rangle} (S^+(i) - S^-(i)) (S^+(j) - S^-(j)), \end{aligned} \quad (41)$$

and

$$\begin{aligned} \mathcal{H}_{odd} = & (J \sin \psi_{ab} + D \cos \psi_{ab}) \sum_{\langle ij \rangle} [S^\zeta(i) S^\eta(j) - S^\eta(i) S^\zeta(j)] \\ & + g_b \mu_B H (\sin \psi_a - \sin \psi_b) \sum_i S^\eta(i) \\ & - \alpha J \sum_{\langle ij \rangle} [S^\zeta(i) S^\eta(j) \sin \psi_a \cos \psi_b - S^\eta(i) S^\zeta(j) \sin \psi_b \cos \psi_a], \end{aligned} \quad (42)$$

where $\psi_{ab} = \psi_a + \psi_b$. The Hamiltonian for the second plane can be obtained from Eqs. (41) and (42) by replacing ψ_a and ψ_b by ψ_c and ψ_d , and changing the sign of D . Finally, the

interlayer coupling is

$$\begin{aligned}
H_{\perp} = & J_{\perp} \sum_{\langle ik \rangle} \left\{ S^{\xi}(i) S^{\xi}(k) - \left[S^{\zeta}(i) S^{\zeta}(k) + S^{\eta}(i) S^{\eta}(k) \right] \cos(\psi_a + \psi_d) \right\} \\
& + J_{\perp} \sum_{\langle jk \rangle} \left\{ S^{\xi}(j) S^{\xi}(k) - \left[S^{\zeta}(j) S^{\zeta}(k) + S^{\eta}(j) S^{\eta}(k) \right] \cos(\psi_b + \psi_c) \right\}. \quad (43)
\end{aligned}$$

It follows from the above equations that the classical ground-state energy is

$$\begin{aligned}
E_0 = & \frac{1}{4} \{ -JS^2 z [\cos(\psi_a + \psi_b) + \cos(\psi_c + \psi_d)] \\
& + \alpha (\sin \psi_a \sin \psi_b + \sin \psi_c \sin \psi_d) - DS^2 z [\sin(\psi_a + \psi_b) - \sin(\psi_c + \psi_d)] \\
& + g_b \mu_B H S (\cos \psi_a - \cos \psi_b + \cos \psi_c - \cos \psi_d) \\
& - 2J_{\perp} S^2 [\cos(\psi_a + \psi_d) + \cos(\psi_b + \psi_c)] \}. \quad (44)
\end{aligned}$$

The minimization conditions with respect to ψ_{μ} yield

$$\begin{aligned}
\psi_a = -\psi_c = & \frac{DSz(g_b \mu_B H - 4J_{\perp} S - \alpha JSz)}{\Delta^2 + 2w^2 - (g_b \mu_B H)^2}, \\
\psi_b = -\psi_d = & -\frac{DSz(g_b \mu_B H + 4J_{\perp} S + \alpha JSz)}{\Delta^2 + 2w^2 - (g_b \mu_B H)^2}. \quad (45)
\end{aligned}$$

When deriving Eqs. (45) we assumed that ψ_{μ} is small, i.e. we consider magnetic fields that satisfy the inequality $g_b \mu_B H \ll \Delta$.

1. Spin gaps in the one-layer model

As we show below, the interlayer coupling almost does not affect the spin-wave gaps. Hence, when calculating the gaps, one may put J_{\perp} to zero, and consider spin waves only in one plane. Performing the Dyson-Maleev transformation of the corresponding terms in the Hamiltonian (41), one gets the bilinear and quartic terms in the Hamiltonian as follows:

$$\begin{aligned}
\mathcal{H}^{(2)} = & \sum_{\langle ij \rangle} \left[JS \left(1 - \frac{\tilde{\psi}_{ab}^2}{2} \right) - DS \tilde{\psi}_{ab} \right] (a_i^{\dagger} a_i + b_j^{\dagger} b_j) \\
& + \sum_{\langle ij \rangle} \left[JS \left(1 - \frac{\tilde{\psi}_{ab}^2}{8} \right) - \frac{DS}{2} \tilde{\psi}_{ab} - \frac{\alpha JS}{2} \right] (a_i b_j + a_i^{\dagger} b_j^{\dagger}) \\
& + \frac{1}{2} \sum_{\langle ij \rangle} \left[\frac{JS}{2} \tilde{\psi}_{ab}^2 + DS \tilde{\psi}_{ab} + \alpha JS \right] (a_i b_j^{\dagger} + a_i^{\dagger} b_j) + g_b \mu_B H \sum_i (a_i^{\dagger} a_i - b_i^{\dagger} b_i), \quad (46)
\end{aligned}$$

$$\mathcal{H}^{(4)} = -J \sum_{ij} \left[\left(1 - \frac{\tilde{\psi}_{ab}^2}{2} \right) a_i^{\dagger} a_i b_j^{\dagger} b_j + \frac{1}{2} \left(1 + \frac{\tilde{\psi}_{ab}^2}{4} \right) a_i^{\dagger} b_j^{\dagger} (a_i^{\dagger} a_i + b_j^{\dagger} b_j) \right]$$

$$\begin{aligned}
& - \frac{\tilde{\psi}_{ab}^2}{8} (a_i b_j^\dagger b_j^\dagger b_j + a_i^\dagger a_i^\dagger a_i b_j) \Big] \\
& + \sum_{ij} \left\{ D \tilde{\psi}_{ab} a_i^\dagger a_i b_j^\dagger b_j - \frac{1}{4} (D \tilde{\psi}_{ab} + \alpha J) [(a_i - a_i^\dagger) b_j^\dagger b_j^\dagger b_j + a_i^\dagger a_i^\dagger a_i (b_j - b_j^\dagger)] \right\}. \quad (47)
\end{aligned}$$

Here $\tilde{\psi}_{ab} = \tilde{\psi}_a + \tilde{\psi}_b$ is the renormalized tilting angle. To calculate it one should add, as in Section III, to the classical ground state energy (44) the quantum correction, i.e. the averaged SW energy, and then minimize the full energy.

The terms linear in the boson operators in Eq. (42) cancel out when the angles given by Eqs. (45) are used. The cubic terms partly cancel out as in the case when H is along the c -axis. The remaining cubic terms are of second order in H and in the other small parameters of the problem, and are therefore irrelevant. Hence, the quantum corrections can be calculated in the Hartree approximation as was explained in the previous Section.

This gives an effective bilinear Hamiltonian, with the coefficients $A_{\mu\nu}$ given by

$$\begin{aligned}
A_{11}(H) &= JSzZ_c \left[1 - R \left(1 + \frac{\xi}{2S} \right) + \frac{\alpha}{2S} \xi \right] + g_b \mu_B H, \\
A_{22}(H) &= A_{11}(-H), \\
A_{12} &= \frac{JSz}{2} \gamma_{\mathbf{q}} Z_m \left[R - \frac{1}{S} (\lambda + 2\eta) + \alpha \right], \\
B_{11} &= -\frac{Jz}{4} [\xi R + 2\eta + \alpha \xi], \\
B_{12} &= JSz \gamma_{\mathbf{q}} Z_c \left[1 - \frac{R}{2} \left(1 - \frac{\xi}{S} \right) - \frac{\alpha}{2} \left(1 + \frac{\xi}{S} \right) \right]. \quad (48)
\end{aligned}$$

Here

$$R = \tilde{\psi}_{ab} \left(\frac{\tilde{\psi}_{ab}}{2} + \frac{D}{J} \right), \quad (49)$$

and the angle $\tilde{\psi}_{ab}$ renormalized by quantum fluctuations is

$$\tilde{\psi}_{ab} = -\frac{2\alpha JD(Z_c S z)^2}{(Z_c \Delta)^2 - (g_b \mu_B H)^2}. \quad (50)$$

Equations (50) and (49) yield

$$R = \frac{\alpha(Z_c \delta)^2 [Z_c^2 \Delta^2 - 2(g_b \mu_B H)^2]}{[Z_c^2 \Delta^2 - (g_b \mu_B H)^2]^2} = \frac{D^2}{2J^2} + O(H^4). \quad (51)$$

Thus, the field dependence of the gaps comes only from the explicit dependence on the field of the matrix elements A_{11} and A_{22} . We get for the in-plane, Ω_{in} , and out-of-plane, Ω_{out} , gaps the following expressions

$$\begin{aligned}
\Omega_{in}^2 &= \frac{1}{2} \left[\tilde{\delta}^2 + \tilde{\Delta}^2 + 2(g_b \mu_B H)^2 - \sqrt{(\tilde{\Delta}^2 - \tilde{\delta}^2)^2 + 8(g_b \mu_B H)^2(\tilde{\delta}^2 + \tilde{\Delta}^2)} \right] \\
\Omega_{out}^2 &= \frac{1}{2} \left[\tilde{\delta}^2 + \tilde{\Delta}^2 + 2(g_b \mu_B H)^2 + \sqrt{(\tilde{\Delta}^2 - \tilde{\delta}^2)^2 + 8(g_b \mu_B H)^2(\tilde{\delta}^2 + \tilde{\Delta}^2)} \right] \quad (52)
\end{aligned}$$

The in-plane frequency Ω_{in}^2 changes sign at the field $g_b\mu_B H = \tilde{\delta}$, when the spin-flop transition takes place.

2. Effect of interlayer coupling

The generalization to a three-dimensional crystal is straightforward. Even though ψ_{ab} now depends on the interlayer coupling w , the quantity R is, as before, equal to $D^2/2J^2$ up to terms of order H^4 . The in-phase gaps therefore do not contain w , and as before they are given by Eqs. (52). The spectrum for spin-waves propagating along the z -axis in small fields $g_b\mu_B H \ll \Delta$ is given by

$$\begin{aligned}\omega_{in}^2(q_z) &= \Omega_{in}^2 + \tilde{w}^2 \left[1 - \frac{4(g_b\mu_B H)^2}{\tilde{\Delta}^2 - \tilde{\delta}^2} \right] (1 \pm c_z), \\ \omega_{out}^2(q_z) &= \Omega_{out}^2 + \tilde{w}^2 \left[1 + \frac{4(g_b\mu_B H)^2}{\tilde{\Delta}^2 - \tilde{\delta}^2} \right] (1 \pm c_z).\end{aligned}\tag{53}$$

Here the upper (lower) sign corresponds to the in-phase (out-of-phase) modes.

B. Field along the a -axis

In this case the field and the DM coupling do not interfere. Hence, the spectrum is the same as one would have for an AF in a field perpendicular to the easy anisotropy axis.²⁵ The only frequencies relevant for the acoustic spin waves in this case are given by

$$\begin{aligned}\omega_{in}^2(q_z) &= \tilde{\delta}^2 + Z_c(g_a\mu_B H)^2 + \tilde{w}^2(1 - c_z), \\ \omega_{out}^2(q_z) &= \tilde{\Delta}^2 + \tilde{w}^2(1 - c_z).\end{aligned}\tag{54}$$

V. DISCUSSION: THEORY VERSUS EXPERIMENT

Consider first the field in the c -direction. The value of g_c in La_2CuO_4 is not well determined yet (see below). We therefore exclude g_c from the expressions for the in-plane gaps, Eqs. (36) and (39), using Eq. (38), and obtain:

$$\begin{aligned}\omega_{in}^2 &= \tilde{\delta}^2 - \tilde{w}^2 \left(\sqrt{H^2/H_c^2 + c_z^2} - 1 \right), \quad H < H_c, \\ \omega_{in}^2 &= \tilde{\delta}^2 + \tilde{w}^2(H/H_c) + \tilde{w}^2(1 - c_z), \quad H > H_c.\end{aligned}\tag{55}$$

These expressions differ from their classical counterparts only by the parameters $\tilde{\delta}$ and \tilde{w} , which should replace their classical values. Thus with the proper choice of the parameter \tilde{w} ($\tilde{\delta}$ and H_c are measured in the experiment) one can fit the experimental data by Gozar *et al.*⁵ in the same way as was done in Refs. 5, 11 and 12. According to Ref. 5, the derivative $d\omega_{in}^2(H > H_c)/dH$ is given by

$$\frac{d\omega_{in}^2(H > H_c)}{dH} = \frac{\tilde{w}^2}{H_c} = 0.35 \frac{(meV)^2}{T}. \quad (56)$$

There is an uncertainty in the value of H_c , because of the hysteresis observed in the first-order WFT. We choose the value $H_c = 6.3$ T, at which the gap ceases to decrease with the increase of the field.⁵ With this value of H_c we obtain from Eq. (56) that $\tilde{w}^2 = 4Z_w^2 J J_\perp = 2.19$ (meV)². Given that $J = 135$ meV,²³ we get:

$$J_\perp = 5.3 \cdot 10^{-3} \text{ meV}, \quad \alpha_\perp = J_\perp/J = 3.9 \cdot 10^{-5}. \quad (57)$$

The ratio \tilde{w}^2/H_c can also be obtained from the relation

$$\tilde{w}^2/H_c = \tilde{\delta} g_c \mu_B (1 - \xi/S), \quad (58)$$

if the g -tensor is known.

According to the calculations performed in Ref. 30, the principal values of the g -tensor are: $g_c = 2.45$, $g_a = g_b = 2.11$. Similar values were later obtained in Ref. 12. The experiment gave somewhat lower values: $g_c = 2.3$, $g_a = g_b = 2.08$,³⁰ or $g_c = 2.24$, $g_a = g_b = 2.06$.³¹ In what follows we use the last values of the g -tensor. Given that $\tilde{\delta}$ is equal to 2.16 meV,⁵ we have: $\tilde{w}^2/H_c = 0.43$ (meV)²/T. This value is somewhat larger than that, which follows from the fit of the field dependence of the gap, Eq. (56). The discrepancy might be caused by the insufficient accuracy of the $1/S$ expansion, since the factor $1 - \xi/S = 1.55$ is significantly larger than one.

With $\tilde{\delta} = 2.16$ meV (2.3 meV according to Ref. 23) we obtain the DM coupling as: $D = \tilde{\delta}/2Z_m = 1.78$ meV (1.90 meV). This value is by a factor two larger than that obtained in Ref. 5. Note that the renormalization of both the in-plane and out-of-plane gaps is given by Z_m rather than Z_c used in Refs. 5 and 23. For more discussion on this subject see Ref. 29.

The dispersion of SW propagating in the c -direction also depends strongly on the field. We have at $H < H_c$

$$\omega_{in}^2(q_z) - \omega_{in}^2(0) = \tilde{w}^2 \left(\sqrt{1 + H^2/H_c^2} - \sqrt{c_z^2 + H^2/H_c^2} \right). \quad (59)$$

The dispersion decreases with the increase of the field, and at the critical field, $H = H_c$, it jumps to the value in zero field,

$$\omega_{in}^2(q_z) - \omega_{in}^2(0) = \tilde{w}^2(1 - c_z). \quad (60)$$

At fields larger than H_c the dispersion does not depend on the field.

When the field is in the b -direction, the measured in-plane gap decreases with the increase of the field as⁵ $\omega_{in}^2 = \tilde{\delta}^2 - \gamma_b H^2$, with $\gamma_b = 0.025$ (meV/T)². This dependence follows from Eq. (52) for fields small in comparison with the out-of-plane gap Δ . In this case we have

$$\gamma_b = \frac{1 + 3(\tilde{\delta}/\tilde{\Delta})^2}{1 - (\tilde{\delta}/\tilde{\Delta})^2} (g_b \mu_B)^2. \quad (61)$$

Given $g_b = 2.06$, $\tilde{\delta} = 2.16$ meV, and the above value of γ_b , we get $\tilde{\Delta} = 5.3$ meV, in good agreement with the neutron measurement result $\tilde{\Delta} = 5.5$ meV.²³ It follows from this result that the out-of-plane anisotropy parameter α is

$$\alpha = \tilde{\Delta}^2 / 8J^2 Z_m^2 = 5.2 \cdot 10^{-4}. \quad (62)$$

The value of α is by an order of magnitude larger than α_{\perp} . This implies that T_N in lanthanum cuprate is determined mainly by the out-of-plane anisotropy.

Finally, we consider the gap when the magnetic field points in the a -direction. The experimental findings were fitted in Ref. 5 by the relation $\omega_{in}^2 = \tilde{\delta}^2 + \gamma_a H^2$, with $\gamma_a = 0.015$ (meV/T)². The theoretical value, which follows from Eq. (54), with $g_a = 2.06$, is somewhat larger: $\gamma_a = 0.0165$ (meV/T)². Note that in the model, which includes ring exchange, Z_c was found to be $Z_c = 0.96$.²⁰ With this value of Z_c one gets $\gamma_a = 0.014$ (meV/T)².

In conclusion, we calculated the effect of quantum fluctuations on the SW spectrum in La_2CuO_4 in an external magnetic field. With the experimental values of the g -tensor, the in-plane gap, and the WFT critical field, a satisfactory agreement of the theory with the experimental findings was obtained. Given the renormalized values of the parameters, which determine the field dependence of the SW gaps, we got new values of the in-plane DM and inter-plane couplings, and of the in-plane anisotropy α .

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APPENDIX

Define the Green's functions

$$G_{\mu\nu}(\mathbf{q}, t) = \langle\langle \alpha_\mu(\mathbf{q}, t), \alpha_\nu^\dagger(\mathbf{q}) \rangle\rangle, \quad F_{\mu\nu}(\mathbf{q}, t) = \langle\langle \alpha_\mu^\dagger(\mathbf{q}, t), \alpha_\nu^\dagger(-\mathbf{q}) \rangle\rangle. \quad (\text{A.1})$$

These functions obey the following equations of motion:

$$\begin{aligned} i \frac{d}{dt} G_{\mu\nu}(\mathbf{q}, t) &= \delta(t) \delta_{\mu\nu} + \langle\langle [\alpha_\mu, \mathcal{H}], \alpha_\nu^\dagger(\mathbf{q}) \rangle\rangle \\ i \frac{d}{dt} F_{\mu\nu}(\mathbf{q}, t) &= \langle\langle [\alpha_\mu^\dagger(-\mathbf{q}), \mathcal{H}], \alpha_\nu^\dagger(\mathbf{q}) \rangle\rangle. \end{aligned} \quad (\text{A.2})$$

Given the Hamiltonian (14), one gets for the Fourier transform of the functions \mathbf{G} and \mathbf{F}

$$\begin{aligned} (\omega \mathbf{I} - \mathbf{A}(\mathbf{q})) \mathbf{G}(\omega, \mathbf{q}) - \mathbf{B}(\mathbf{q}) \mathbf{F}(\omega, \mathbf{q}) &= \mathbf{I}, \\ (\omega \mathbf{I} + \mathbf{A}(\mathbf{q})) \mathbf{F}(\omega, \mathbf{q}) + \mathbf{B}(\mathbf{q}) \mathbf{G}(\omega, \mathbf{q}) &= 0, \end{aligned} \quad (\text{A.3})$$

where \mathbf{I} is a unit matrix.

The equations of the eigenvalue problem can be written as

$$\begin{aligned} (\omega \mathbf{I} - \mathbf{A}) |G\rangle - \mathbf{B} |F\rangle &= 0, \\ (\omega \mathbf{I} + \mathbf{A}) |F\rangle + \mathbf{B} |G\rangle &= 0, \end{aligned} \quad (\text{A.4})$$

where $|G\rangle$ and $|F\rangle$ are columns of the corresponding matrices.

This gives

$$\begin{aligned} \omega |G - F\rangle &= (\mathbf{A} + \mathbf{B}) |G + F\rangle, \\ \omega |G + F\rangle &= (\mathbf{A} - \mathbf{B}) |G - F\rangle. \end{aligned} \quad (\text{A.5})$$

Hence,

$$\omega^2 \mathbf{I} - (\mathbf{A} + \mathbf{B})(\mathbf{A} - \mathbf{B}) = 0. \quad (\text{A.6})$$

Thus, the squares of the spin-wave energy, ω^2 , are the eigenvalues of the matrix¹⁹

$$\mathbf{M} = [\mathbf{A}(\mathbf{q}) + \mathbf{B}(\mathbf{q})] \times [\mathbf{A}(\mathbf{q}) - \mathbf{B}(\mathbf{q})]. \quad (\text{A.7})$$

The quantities η and λ , defined in Eq. (28), are related to the Green's functions as

$$\begin{aligned}\eta &= \frac{1}{\pi} \int d\omega \Im \sum_{\mathbf{q}} G_{21}(\omega, \mathbf{q}) \gamma_{\mathbf{q}}, \\ \lambda &= \frac{1}{\pi} \int d\omega \Im \sum_{\mathbf{q}} F_{11}(\omega, \mathbf{q}).\end{aligned}\tag{A.8}$$

Together with Eqs. (A.3), this leads to the relations

$$\begin{aligned}\eta &= -\frac{A_{12}}{2JSz} \sum_{\mathbf{q}} \frac{\gamma_{\mathbf{q}}^4}{(1 - \gamma_{\mathbf{q}}^2)^{3/2}}, \\ \lambda &= \frac{A_{12}}{2JSz} \sum_{\mathbf{q}} \frac{\gamma_{\mathbf{q}}^2}{(1 - \gamma_{\mathbf{q}}^2)^{3/2}}.\end{aligned}\tag{A.9}$$

Thus, we have

$$\eta + \lambda = \frac{A_{12}}{JSz} \xi.\tag{A.10}$$

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